JOURNAL OF COMPLEMENTARY MEDICINE RESEARCH, 2020
VOL 11, NO. 1
10.5455/jcmr.2020.11.01.28

RESEARCH ARTICLE

Open Access

Geochemical variables and fluoride ion concentration levels impart infectious diseases: A systematic study of fluoride ion removal using natural adsorbents at low cost

B. Vijila and J.Joseph[*]
Department of Chemistry, Noorul Islam Centre for Higher Education, Kumaracoil-629180, Tamil Nadu

ABSTRACT

In this proceedings under way was concentrated on water collection samples in Tirunelveli district from well (open and bore well), tape, river and the pond. The results were characterised by physico-chemical and microbiological studies. Depending on the amount fluoride has both beneficial and adverse effects on human health. The allowable limit for fluoride in drinking water is 1.5 mg / L according to India's and the World Health Organization 's National Standards for Drinking Water Quality. Beyond this cap, fluoride can lead to a number of diseases including skeletal and dental fluorosis, brittle bones, cancer, infertility, brain damage, Alzheimer's syndrome and thyroid disease. Fluoride removal from the sample of drinking water is important for utility purposes. Among the literature methods, the adsorption method is deemed more suitable for defluoridation due to its usability, efficacy and economic viability. In these present investigations, the low cost agricultural materials as adsorbents were utilized for fluoride removal and also compared the efficiency. The experimental observations are discussed. We conclude that numerous human activities in the soil deliberate the release of toxic ion levels, which can lead to serious human health consequences for the environment. We recommend that the public health sector has taken appropriate action and organized more awareness camp related to health issues with the use of natural adsorbent materials for purification and removal as well.

ARTICLE HISTORY

Received February 7 2020, Accepted March 4 ,2020 Published July 11, 2020

KEYWORDS

fluoride,charcoal, adsorption

INTRODUCTION

This current paper reported that the consumption of ground and surface water without any treatments [1]. Hunter P.R et.al reported that arsenic, mercury, cyanide and fluoride are present high amount in groundwater [2]. Human activities, environmental factors and characteristics of geographical studied area depend on the ground water safety measures. Fawell J et.al has been reported ingestion of contaminated ground and surface water causes water-borne diseases [3]. Water quality was confirmed by physico-chemical and biological constituents for drinking purpose ground water is consider as mainly useful resource to us. Maximum acceptable fluoride concentration is 1.5mg/L reported by WHO [4]. If the fluoride concentration is increase, causes fluorosis disease.

In Tamil Nadu, population density is increased by day to day. Population increases means contamination of

[*] **Contact**: B. Vijila and J.Joseph Department of Chemistry, Noorul Islam Centre for Higher Education, Kumaracoil-629180, Tamil Nadu

water, air and soil also increases. This evidences water is one of the apparent victim of pollution.

In India, Tirunelveli is high populated district in Tamil Nadu state. Due to high population the water sources are contaminated by diverse of pollutants. Nowadays, fluoride is the important pollutant present in water sources. Fluoride concentration increase causing teeth mottling, kidney and thyroid gland damage [5].

Adsorption, electro - coagulation, coagulation – precipitation, ion exchange and membrane separation are diverse techniques involved this defluorination [6-10]. Different techniques are involved for this fluoride removal contamination but adsorption method is most favourable, because of its easiest method and low cost.

Powdered activated carbon usage shows good results for fluoride reduction at pH 3 or less[11]. Muhammad et.al, reported that fluoride removal from polluted water using bone char, synthesized with thermal pyrolysis of cattle bones [12]. Srimurali et.al reported, batch adsorption studies are used for eliminating fluoride from low cost water products [13].

In this present investigation, the quantity of fluoride pollution in different sources of drinking water and different techniques to be adopted for removing fluoride from water, especially through adsorption using different charcoals, were based.

Experimental & Methods

Study area

Tirunelveli is one of Tamil Nadu's largest districts spread over 6759 square kilometers of area. It is put on earth as longitude 77°.05' to 78°.25' in the east and 8°.05' to 9°.30' as latitude in the north. Thirunelveli means Sacred Paddy Hedge. Kurunchi, Mullai, Marutham and Neithal are the four land structures of Tirunelveli district. Tamiraparani river is a perennial river, which is the salvation of this district and feeds drinking water to the district.

In Tirunelveli district, water is polluted by various contaminants such as municipal wastes, drainage and agricultural wastes. Antrophogenic activities also affect the water bodies. Peoples having there is no awareness about water treatment and waste water reusage. Waste water and contaminants are directly entering into the water bodies caused waterborne diseases. The use of insecticides and pesticides also contaminates the water sources in agricultural land.

Sample collection

Water samples gathered from pond, tape, well (open, bore) and river. Clean stoppered bottles are used without air bubbles, for this sample collection. Before sampling the stoppered bottles were rinsed thoroughly and tightly closed after sampling then sealed. Depending upon the physical qualities (colour, odour and taste) water samples collected, for this physical and chemical analysis.

Analysis of water sample

Colour, taste, odour, temperature, pH, turbidity and electrical conductivity are the physical parameters. The chemical parameters are acidity, alkalinity, total hardness, total dissolved solids, chloride, calcium, magnesium, nitrate, sulphate, iron, ammonia, and fluoride.

Determination of water quality parameters

pH meter is used to measured the pH value of samples. The total dissolved solids were determined by using Gravimetric method. Hardness of water was determined by EDTA titration. Volumetric titrations performed for estimating the total alkalinity of water. Nitrite content by qualitatively and quantitative tests applicable only for abnormal amount samples present in it.

Thiocyanante or colour comparison method was performed to estimate iron content. Nitrate was also estimated by colour comparison method by preparing standard nitrate solutions and thereby comparing it with the colour given by both sample and standard nitrate solution on addition of 2,4-phenol disulphonic acid in alkaline medium. Bacteriological analysis were done by calculating standard plate count where bacteria where allowed to develop colonies on nutrient agar medium and by multiple tube method using Mac Conkeybroth.

Chloride content determined by argentometric titration and ammonia by colorimetric method. Turbidity and sulphate content were determined by Nepheloturbidimetric method.

Table 1: Physico-Chemical parameters bore well samples (No.1-5) and open wells (No.6-8)

Sl.No	Parameters	1	2	3	4	5	6	7
1	Turbidity	8.2	0.2	0.2	0.1	0.1	16.3	0.1
2	Taste, Colour, Odour	objectionable Taste	objectionable Taste, Colour and odour	objectionable Taste	objectionable Taste	objectionable Taste	objectionable Taste, Colour and odour	objectionable Taste
3	Temperature	32.3	33.4	32.8	33.1	31.7	31.6	32.8
4	pH	6.45	7.24	5.84	6.44	5.10	6.53	5.98
5	E C	169.4	285.0	127.8	234.2	189.4	244.0	179.0
6	T D S	118.3	191.7	85.5	153.4	126.6	164.5	117.3
7	Acidity	26.0	12.0	22.0	19.0	38.0	14.0	20.0
8	Alkalinity	43.0	68.0	20.0	52.0	18.0	72.0	26.0
9	Total Hardness	37.0	45.0	29.0	73.0	32.0	43.0	38.0
10	Calcium	4.02	8.13	3.68	5.34	4.81	8.02	5.63
11	Magnesium	3.67	5.73	3.59	5.43	4.36	5.83	4.67
12	Chloride	124.0	62.0	40.0	43.0	66.0	38.0	52.0
13	Fluoride	1.82	2.07	1.40	1.008	0.98	1.83	1.37sss
14	Iron	1.82	0.01	3.54	0.25	0.01	0.04	0.01
15	Nitrate	BDL	BDL	5.06	BDL	43.73	BDL	8.23
16	Sulphate	23.0	BDL	BDL	BDL	BDL	BDL	BDL
17	Ammonia	0.63	0.01	0.32	BDL	0.1	BDL	0.03

BDL-Below Desirable Limit

Results & discussion

The physico-chemical parameters directly related to the safety of the drinking water to human consumption. The samples are collected randomly and about 28 samples were physically analysed. Out of these 28 samples, 9 from Bore wells, 6 from open wells, 8 from ponds, 3 from Tapes and 2 from rivers. During the physico-chemical analysis, about 14 samples were polluted with physical contaminants (excess turbidity, pungent odour, change in colour and taste). Then these 14 samples undergo fluoride detecting test. After this analysis, it concluded that 13 samples polluted with fluoride contaminants (samples 5 from Bore wells, 3 from open wells, 4 from ponds and 1 from river). Finally, the samples were analysed and the According to the charts of IS 10500:2012 released by the Bureau of India, water quality norms followed. Analytical grade reagents were used and distilled water is used for solution preparation.

Temperature

In an aquatic atmosphere temperature is an important parameter for its effects on the chemical and biological reactions in living organisms. The atmospheric temperature differs in various locations,

showing some desirable changes in the water temperature. Groundwater temperature and atmospheric temperature are differing due to high specific heat of water. Temperature variation in studied area is 15 to 35°C.

Colour

Generally, colour of the samples was classified into true and apparent colour. A decayed Organic component (grass, dead leaves, etc) gives the true colour. This was seen in surface water. Decaying of inorganic elements (copper, iron, manganese, etc) gives the apparent colour of the water.

Decaying organic and inorganic components present in the water appeared as visible. WHO reported that the maximum permissible range is <15 TCU. Water samples with higher colour concentration not suitable because its need more water treatment.

Table 2: Physico-Chemical parameters of pond samples (No.9-12) and river water (No.13)

Sl.No	Parameters	8	9	10	11	12	13
1	Turbidity	8.2	0.2	0.2	0.1	0.1	16.3
2	Taste, Colour, Odour	objectionable Taste and Colour	objectionable Taste, Colour and odour	objectionable Taste	objectionable Taste	objectionable Taste	objectionable Taste, Colour and odour
3	Temperature	32.3	33.4	32.8	33.1	31.7	31.9
4	pH	6.45	7.24	5.84	5.97	5.10	6.53
5	E C	153.8	286.0	127.8	177.0	190.4	247.0
6	T D S	112.6	191.6	85.6	118.3	127.6	165.5
7	Acidity	16.0	4.0	21.0	19.1	38.0	14.0
8	Alkalinity	24.3	67.0	20.0	25.9	17.0	62.0
9	Total Hardness	63.0	43.0	28.0	31.0	39.0	47.0
10	Calcium	4.33	8.62	4.48	5.59	4.51	8.12
11	Magnesium	5.24	5.13	3.83	4.23	4.85	5.73
12	Chloride	57.0	62.0	40.0	51.0	64.0	36.0
13	Fluoride	1.23	1.04	1.7	2.64	1.02	1.53
14	Iron	0.02	0.01	0.14	0.02	0.19	0.01
15	Nitrate	4.89	BDL	5.07	8.43	46.72	BDL
16	Sulphate	BDL	BDL	28.0	BDL	BDL	BDL
17	Ammonia	BDL	0.01	BDL	0.02	BDL	0.02

BDL-Below Desirable Limit

Table 3: Water Quality Parameters as per IS 10500:2012

Sl.No	Parameters	Unit	Desirable Limit	Permissible Limit
1	Turbidity	NTU	1	5
2	Taste, Colour, Odour		Unobjectionable	Unobjectionable
3	Temperature	^0C		
4	pH		6.5-8.5	6.5-8.5
5	Electrical Conductivity	$\mu mho/cm^2$		
6	Total Dissolved Solids	mg/L	500	2000
7	Acidity	mg/L		
8	Alkalinity	mg/L	200	600
9	Total Hardness	mg/L	200	600
10	Calcium as Ca^{2+}	mg/L	75	200
11	Magnesium as Mg^{2+}	mg/L	30	100
12	Chloride	mg/L	250	1000
13	Fluoride	mg/L	1	1.5
14	Iron as Fe^{2+}	mg/L	0.3	No Relaxation
15	Nitrate	mg/L	45	No Relaxation
16	Sulphate	mg/L	200	400
17	Ammonia	mg/L	0.5	No Relaxation

Turbidity

The turbidity analysis of water samples reveals the transparency. It is mainly influenced by the constituents present in sample. Turbidity also makes the water aesthetically unacceptable. Acceptable value is below 5 NTU reported WHO. In the present study, many of the samples were attained within the desirable limit of 1 NTU. Iron and suspended organic components are the main factors for turbidity.

pH

The pH intends the strength of samples acidity or alkalinity. The amount of dissolved CO_2 present in the sample gives the pH value. WHO and BIS were

reported, 6.5 – 8.5 is the acceptable and safe range for drinking water,

Chloride
250mg/L is the permissible chloride limit in the drinking water and the concentration was present in desirable limit.

Iron
0.3 mg/L is the desirable limit of iron in drinking water. Excess of iron concentration in drinking water causes bad taste, colour change, brown precipitate, staining on tiles, cloths and sink. After the iron removal the samples were fit for drinking purpose.
In the present study, samples 1, 3 not suitable for drinking purpose, which needs some treatment to reduce the iron concentration.

Calcium
About 1.2 kg of calcium present in human body. It is a nutritional mineral. Ingestion of excess calcium causes health problems such as impaired kidney function and hyper calcemia.

Magnesium
Magnesium is the important component for hardness of water. Deficiency of magnesium produces, decreases in intracellular potassium concentration and increases calcium concentration in human body. In this study, magnesium is not responsible for hardness producing.

Table 4: BOD levels and their water quality

BOD level (mg/L)	Water quality
1-2	Excellent
3-5	Good
6-9	Poor- Polluted
More than 10	Very poor- Highly contaminated

Table 5: Effective removal of different charcoals

	Amount of charcoal used in (g)	Amount of fluoride present in (mg/L)	Amount of fluoride after filtration in (mg/L)	% of removal of fluoride
Wood charcoal	10	2.64	1.65	37.5
	15	2.64	1.24	53
	20	2.64	0.74	72
Activated charcoal	10	2.64	2.48	6
	15	2.64	2.31	12.5
	20	2.64	1.98	25
Rice husk	10	2.64	0.74	71.8
	15	2.64	0.44	84.3
	20	2.64	0.08	96.8
Animal charcoal	10	2.64	2.56	3.1
	15	2.64	2.48	6.25
	20	2.64	2.06	21.8

Nitrate
Nitrate is one of the significant parameter in this analysis. The desirable limit of nitrate is 45 mg/ L and no relaxation for permissible limit. Presence of excess nitrate in drinking water may cause blue baby syndrome.

Sulphate
250 mg/L is the maximum permissible limit of sulphate in drinking water, reported by WHO. Unpleasant rotten egg odour and bitter taste caused by water contain excess sulphate content.

Acidity
Acidity is often correlated with low pH. No definition for acidity is provided when pH is maintained in drinking water as 6.5-8.5. Many of the water samples in the present study showed low pH value. Such samples were also found to be extremely acidic in nature. Acidic water has adverse reacted to metals, utensils, cloths, etc.

Total Hardness and Alkalinity
Total hardness of water is produced by the presence of Calcium and magnesium salts of carbonates,

chlorides and sulphates. 600 mg/L is the maximum tolerable limit of total hardness of drinking water. In this present study, alkalinity and total hardness of all samples not exceed the desired 200 mg / L limit.

Dissolved Oxygen (DO)

The oxygen dissolved is an important element for both aerobic and anaerobic organisms. The optimum DO level is 6 mg / L for meaningful fish growth and other aquatic life. In this analysis, the DO was observed in the permissible limits within the sampling area. Hence the sample of water is safe for marine life.

Biological Oxygen Demand (BOD)

BOD is the amount of oxygen essential in aerobic conditions for the decomposition of dissolved organic substance. Determining the strength of pollution, the nature of sewage and the type of effluent present is crucial technique. A high BOD level indicates the presence of higher quantities of micro-organisms that deliberately increase pollution levels in water bodies.

Fluoride

Fluoride is naturally found in earth crust. In this study area may be derived from sediments which originated from fluorspar (fluorite), apatite, topaz and cryolite which forms part of the aquifer material. The breaking of rocks or extraction of compounds from rock is utilized to produce phosphate containing fertilizer and deposited into soil. In our human body, fluoride present in the form of calcium fluoride. This is found in teeth and bones.

In drinking water the desirable limit for fluoride is 1mg / L. Only 2 out of 14 samples showed below 1mg/L and the remaining 12 samples showed above the desired limit. It is evident in this analysis that the sample contains a small concentration of fluoride (below 1mg / L) which causes a change in taste. Therefore, to consumers it is not edible.

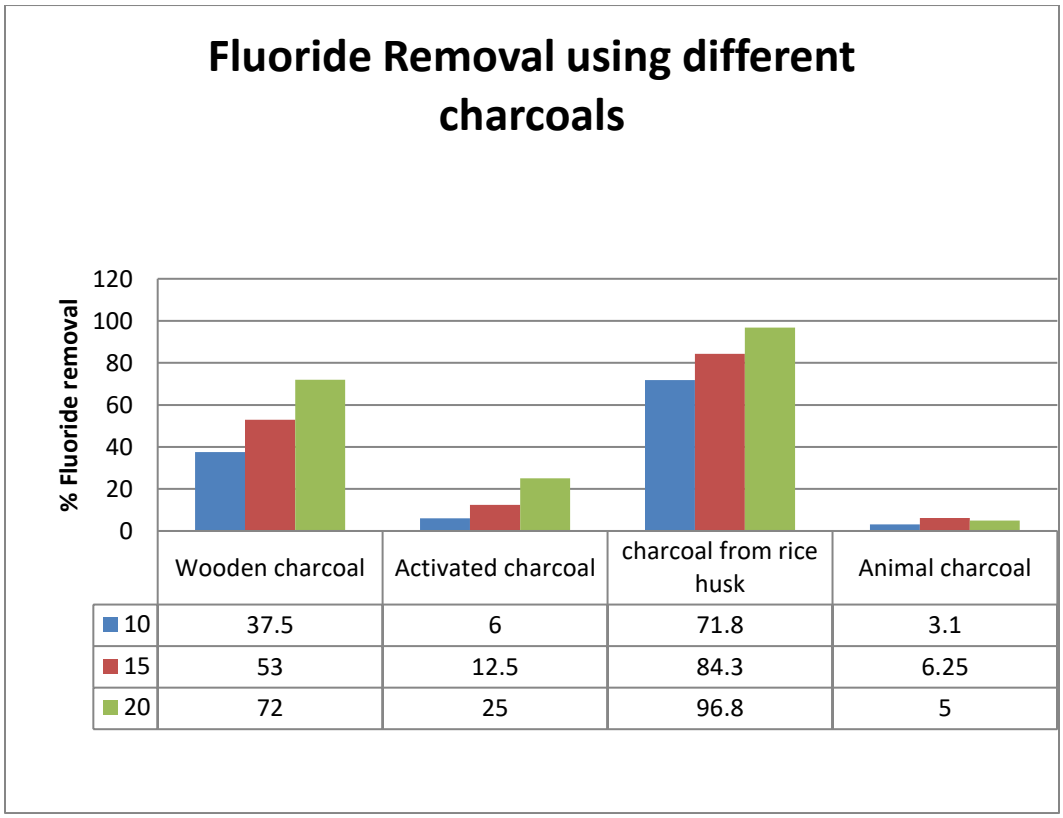

Figure1: Fluoride removal using different charcoals

REMOVAL OF FLUORIDE USING DIFFERENT CHARCOALS

Activated carbon is a versatile material that encompasses structural architecture with large surface area, porous, material regeneration and adsorption capability etc. Coal with a surface enriched with natural carbon with a high surface area that stimulates researchers to extract pollutants from

water supplies. The pollutants of the water cause different kinds of diseases for the men. Therefore the need for proper selection of carbon enriched activated carbon from natural origin to eliminate possible pollutants from water bodies is growing. In the present study, the different charcoal was used for the removal of fluorides (i) Wood charcoal (obtained from , wood charcoal is prepared by char the coconut shell); (ii) Activated charcoal, (iii) charcoal from rice husk and iv) animal charcoal. The composition of animal charcoal consists of less than 10 per cent of carbon and a higher percentage of calcium phosphate and a minimum of other calcium salts. The oxygen on the surface that contains molecules is very small and it affects the ability of adsorption. It also has lower surface area relative to activated carbons. The charcoal from Wood is hydrophilic and has an oxidised surface. Due to its abundance and low cost the rice husk is widely used. The rice husk, because of its cellular structure skeleton, possesses carbon enriched by high porous and large surface area. The four separate charcoals were filled in burettes in the present investigations as 10 g, 15 g, and 20 g. Then added 25 ml of water that contains maximum sample number 11 fluorides, the fluoride content is 2.64. When the charcoal has been filtered again tested for fluoride and the results are reported.

From the table and graph, it is clear that the raise husk charcoal produced a spectacular fluoride removal test. Total fluoride reduction was achieved by passing water through rice husk charcoal (20 g) and is approximately 96.8%. The experimental observations showed that the amount of fluoride removal increases as the adsorbent dosage increases.

Conclusion

This current investigation was persistent on the analysis of various water sources and fluoride contamination removal. Water samples were collected from different places and analysed by using standard methods. Water sources were contaminated by various pollutants, fluoride is one of the important pollutants present in the water sources. If, fluoride is present that will affect the properties and quality of water such as colour, odour and taste of the water.

A safe and efficient charcoal was used in the adsorption process to extract the fluoride from the water sample. Widespread, charcoal is used in water filters to eliminate dissolved gases. Activated and wood charcoals are primarily used in water filters with low adsorption. From this study, the adsorption rate of rise husk charcoal is higher against fluoride and about 96%. The surface area, volume of pore and morphological surface characteristics of charcoal materials define the types of molecules that could be adsorbed onto it. Charcoal's core surface expresses the positive charged group which attracts negative fluoride charges through the phenomenon of chemisorptions (electrostatic interaction).It is concluded that the performance of various analysis indicated that the sample collected from Tirunelveli district is well suited for human consumption, agricultural productivity, industrial processes whereas some sample needs appropriate treatments for the removal of contaminants before utility.

References

1. Edokpayi, J.N., Rogawski, E.T., Kahler, D.M., Hill, C.L., Reynolds, C., Nyathi, E., Smith, J.A., Odiyo, J.O., Samie, A.; Bessong, P., et al. Challenges to Sustainable Safe Drinking Water: A Case Study of Water Quality and Use across Seasons in Rural Communities in Limpopo Province, South Africa, *Water* , 2018,10, 159.
2. Hunter, P.R., MacDonald, A.M., Carter, R.C., Water Supply and Health, *PLoS Med.* , 2010,7, 1000361.
3. Fawell, J., Niewenhuijen, M.J, Contaminants in drinking water: Environmental pollution and health, Br. Med. Bull. 2003, 68, 1–5.
4. A.Goswami,M.K.Purkait, ' Removal of fluoride from drinking water using nanomagnetite aggregated Schwermannite' *Journal of Water Process Engineering* , 2014,1, 91–100.
5. Krishna Kumar Yadavb, Sandeep Kumarc, Quoc Bao Phamh, Neha Guptab, Shahabaldin Rezaniad, Hesam Kamyabe, Shalini Yadavf, Jan Vymazalg, Vinit Kumarb, Doan Quang Tria,*, Amirreza Talaiekhozanii, Shiv Prasadc, Lisa M. Reecej, Neeraja Singhk, Pradip Kumar Mauryal, Jinwoo Chod, 'Fluoride contamination, health problems and remediation methods in Asian groundwater: A comprehensive review' *Ecotoxicology and Environmental Safety* , 2019, 182, 109362.
6. N.K Mondal, R. Bhaumik, P. Roy, P. Das, J.K. Datta, Investigation on fixed bed column performance of fluoride adsorption by sugarcane charcoal, *J. Environ. Biology*., 2013, 34,1059-1064.
7. S. Singh, B. Kumar Singh, Removal of fluoride from groundwater by carbonised Punica granatum carbon ("CPGC") bio-adsorbent, *J. Geosci. Environ. Protec.*, 2015, 3, 1-9.
8. Nawlakhe, W., Paramasivam, R., 'Defluoridation of potable water by Nalgonda technique' *Curr. Sci.* 1993, 65, 743–748.
9. Z. Amof, B. Barioub, N. Mameri, M. Taky, S. Nicolasb, A. Elmidaoui, Fluoride removal from

brackish water by electrodialysis. *Desalination*, 2001, 133, 215-223.

10. E. Bazrafshan, K. A. Ownagh, A. H. Mahvi, 'Application of electro coagulation process using Iron and aluminum electrodes for fluoride removal from aqueous environment' *E. J. Chemistry,* 2012,9, 2297-2308.
11. R. Mckee, and W.S. Johnston, 'Removal of fluorides from drinking water using low-cost adsorbent', *Indian Journal of Environmental Health*, 1999, 41 (1), pp. 53– 58.
12. Muhammad Kashif Shahid, Jun Young Kim, Young-Gyun Choi, Synthesis of bone char from cattle bones and its application for fluoride removal from the contaminated water, *Groundwater for Sustainable Development* ,2018.
13. M.SrimuraliA. PragathiJ. Karthikeyan, A study on removal of fluorides from drinking water by adsorption onto low-cost materials, *Environmental Pollution*, 1998, Volume 99, Issue 2, Pages 285-289.